RAPID COMMUNICATION

High critical currents by isotropic magnetic-flux-pinning centres in a 3 μm-thick YBa₂Cu₃O₇ superconducting coated conductor

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Abstract

The critical current densities $J_{c-w}(H)$ per unit width of the tape were measured at 75 K in applied magnetic fields H on a 3 μ m-thick YBa₂Cu₃O₇ film on a metallic substrate fabricated using the BaF₂ process. $J_{c-w}(H)$ was the highest reported for YBa₂Cu₃O₇ conductors with H perpendicular to the tape, and was independent of the angle between H and the tape plane for $\mu_0 H \leq 3$ T. The film did not contain artificial flux-pinning centres, implying the existence of a strong isotropic-pinning centre in undoped YBa₂Cu₃O₇ films, thus opening up unexplored avenues for improving performance of coated conductors.

Superconducting coated conductors—flexible metallic tapes coated with films of the superconductor YBa2Cu3O7 (YBCO)—are rapidly progressing toward practical implementation in public electric utilities [1]. Further increases in their current-carrying capacity J_{c-w} , expressed as amperes per millimetre width, of these tapes are needed to improve performance of these devices and to make them viable products [2]. A requirement for achieving high J_{c-w} is synthesis of a thick ($\gg 1 \mu m$) YBCO layer with high critical current density J_c (A mm⁻²), since J_{c-w} is equal to the product of J_c and film thickness. However, combining large thickness and high J_c in a YBCO layer is very difficult [2, 3]. A key obstacle is the incorporation of strong flux-pinning centres without degrading structure of the thick YBCO film. Some films with excellent J_{c-w} were recently made by forming a multilayer of YBCO and CeO₂ [4], and by introducing small, correlated lattice defects in the YBCO films by adding BaZrO₂ [3]. More recently, it was shown that single layer YBCO films as thick as 4.8 μ m could be made to carry either high J_{c-w} of $\sim 100 \text{ A mm}^{-1}$ in self-field (i.e. H = 0) but with strongly decreasing $J_{c-w}(H)$, or somewhat lower J_{c-w} in self-field but with higher $J_{c-w}(H)$,

depending on the conditions of the top buffer layer between the metal substrate and the YBCO [5]. Here, H is applied magnetic field.

These YBCO layers were deposited by the pulsed laser deposition (PLD) technique, which is considered by many to be uneconomical for commercial conductor fabrication. We recently demonstrated [6] that c-axis-oriented 3 and 4 μ m-thick YBCO films could be made on a metallic substrate by the BaF2 process [7, 8], which is a commercially scalable process. These 3 \times 10 mm² coupons sustained very high J_{c-w} in a 1 T perpendicular field at 77 K of 11.4 and 15.2 A mm¹ for the 3 and 4 μ m-thick films, respectively. In addition, the critical temperature T_c of a 3 μ m-thick film was 92.5 K with a transition width of 0.5 K (measured using a resistance method).

These $3 \times 10 \text{ mm}^2$ tape coupon specimens were synthesized using the following processing sequence. Precursor layers were deposited by vacuum co-evaporation of Y, BaF₂ and Cu on textured Ni–W alloy tapes [9] buffered by oxide layers³. The composition of the precursor film was intentionally

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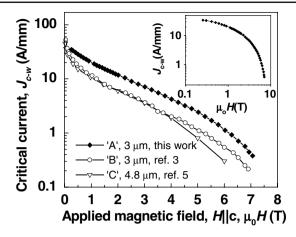


Figure 1. Critical current densities $J_{c-w}(\mu_0 H)$ for the field perpendicular to the film of the present 3 μ m-thick film ('A', this work) are compared with those for the PLD films of the 3 μ m-('B', reference [3]) and 4.8 μ m-thick ('C', reference [5]) films which contained artificial correlated pinning centres. $J_{c-w}(\mu_0 H)$ for samples 'A' and 'B' was measured at 75.5 K while that for 'B' was measured at 77 K. The inset shows the same data for the present film plotted in a log-log plot to illustrate that $J_{c-w}(H)$ does not fall off as $H^{-\alpha}$ where α is a constant.

made Y rich, Y = 1.2, to suppress the formation of undesirable phases such as BaCuO₂. The YBCO conversion process was carried out in a processing gas at sub-atmospheric pressure [6, 8]. During conversion, YBCO grew epitaxially on the substrate at a rate of \sim 0.7 nm s⁻¹ with the crystallographic *c*-axis perpendicular to the tape plane. This growth rate was very fast for the BaF₂ process, with typical rates being \sim 0.1 nm s⁻¹ at atmospheric pressure [10]. The local growth rate of the BaF₂ process is lower than typically achieved in a large PLD system; however, the BaF₂ process has the advantage of a very large processing area, which is only limited by the size of the furnace, thus making high volume production possible.

In addition to the above-mentioned tapes with very high critical current densities J_{c-w} , 25 coupons of the similarly processed 3 and 4 μ m-thick films have been evaluated at 77 K in external magnetic fields up to 1 T. All of these samples demonstrated $J_c > 10^6$ A cm⁻² in self-field and very strong retention of J_c in a magnetic field with an average ratio of $J_c(0 \text{ T})/J_c(1 \text{ T}, H \parallel c) = 3.2 \pm 0.2$. Hence, we concluded that these specimens deserved a detailed study in higher magnetic fields.

For this purpose, a 3 μ m-thick and patterned specimen from the earlier study was selected. The size of the bridge was 0.24 mm wide and 2 mm long. The self-field J_{c-w} of this bridge was \sim 70 A mm⁻¹ at 77 K. A detailed study of the infield behaviour of the critical currents I_c in $\mu_0 H$ up to 7 T was carried out at Los Alamos National Laboratory. In addition to the measurements of I_c with the fields applied perpendicular to the tape plane, these experiments included measurements of $I_c(H,\theta)$ where the external magnetic fields were rotated from the angle $\theta=0^\circ$ to 90° . In this case θ is the angle between the field direction and the tape plane normal. The magnetic field was rotated in a plane perpendicular to the direction of the transport current. At $\theta=0^\circ$, the field direction was parallel to the c-axis of the YBCO layer, $H \parallel c$, and at $\theta=90^\circ$,

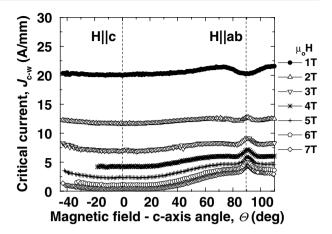


Figure 2. $J_{c-w}(\mu_0 H, \theta)$ for $\mu_0 H$ from 1 to 7 T is plotted as a function of the angle θ between the plane normal to the film and the direction of the applied field. Note the practically isotropic behaviour of $J_{c-w}(\mu_0 H, \theta)$ in fields below 3 T.

the field was parallel to the a-b plane of the YBCO layer, $H \parallel ab$. During the measurements, the sample was immersed in liquid nitrogen, which boiled at 75.5 K at the elevation of Los Alamos, NM. To determine the critical current, we used a standard four-probe technique and an electric field strength criterion of $0.1~\mu \rm V~mm^{-1}$.

Figure 1 presents the critical current densities $J_{c-w}(H)$ for the $H \parallel c$ orientation of the 3 μ m-thick film (sample 'A' in figure 1) and these are compared with those for two thickfilm conductors, which represent recent advances of the films made by PLD. One reference sample, labelled 'B' in figure 1, was a 3 μ m-thick layer which was deposited by pulsed laser ablation of a YBCO target doped with BaZrO₃ (BZO) [3]. The lattice mismatch between the YBCO matrix and BZO forced BZO particles to align into a dense array of quasi nano-rods. These nano-rods grew from the substrate through the bulk of the YBCO film to the film surface [3]. The fabrication of sample 'C' in figure 1 followed a different strategy for creating extended defects in a 4.8 µm-thick YBCO film deposited by PLD. In this case nanometre-sized out-growths of SrTiO₃ on the substrate surface served as nuclei for dislocations, which propagated through the whole film in a direction normal to the film surface [5].

It is generally accepted, that correlated defects such as these provide the strongest flux pinning while the magnetic field direction is parallel to them [3, 5, 11]. However, as shown in figure 1, the critical current densities $J_{c-w}(H)$ of our 3 μ m-thick film exceed those of the samples 'B' and 'C' by a factor of \sim 2 in fields over 0.1 T in an orientation $H \parallel c$ which is the most favourable field direction for the above films with correlated pinning centres. Since our film does not incorporate any intentionally added alloying elements nor correlated pinning centres, the high values of $J_c(H)_{c-w}$ of the film is quite intriguing. The angular dependence of the critical currents, shown in figure 2, provides a better insight into the nature of the pinning in our films. Three observations are important: (a) at low fields, e.g. $\mu_0 H < \sim$ 3 T, $J_{\mathrm{c-}w}(\mu_0 H)$ is nearly independent of θ ; (b) the angular dependence at high fields is primarily due to the electronic mass anisotropy of YBCO; and (c) the very weak $J_{c-w}(\theta)$ peak at $\theta = 0^{\circ}$

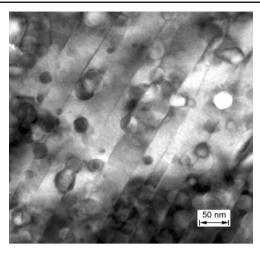


Figure 3. A typical plane-view TEM image for a similarly processed YBCO film on a buffered Ni–W substrate. Circular precipitates are Y_2O_3 and the parallel lines are the twin boundaries.

indicates that there is no significant correlated pinning along the c-axis. Based on these observations, it can be concluded that the pinning centres are isotropic and are not correlated or aligned to any particular direction. Such strong, isotropic flux pinning has not been previously observed in coated conductors at liquid nitrogen temperatures.

In addition, it is interesting to note that the power-law dependence of J_c on H reported and often observed previously for YBCO films [12] is not found here, as shown in the inset of figure 1. This may possibly be a signature for near elimination of the weak links to transport currents at misoriented grain boundaries in a YBCO film. This is supported by the observation of the magnetization peak in this film occurring at H = 0 rather than in the first or third quadrant of a magnetization curve [13].

The pinning centres responsible for the dramatic improvement in our film's properties remain difficult to Transmission electron microscopy (TEM) of similarly processed films was performed. In figure 3, a typical plane-view image of the film is shown. In the figure, the only clearly distinguishable candidates for flux-pinning centres are circular precipitates of Y₂O₃ 10-40 nm in diameter and the parallel twin boundaries. The density of Y_2O_3 particles is estimated to be $4 \times 10^{21} \text{ m}^{-3}$ assuming the thickness of the TEM specimen is 50 nm, which is consistent with the precursor being Y rich. Interestingly, this precipitate density is very close to the 4.8×10^{21} m⁻³ estimated using the same assumption for the TEM specimen thickness for a 0.12 μ m YBCO film, which was made using a similar process and substrate [14]. Despite the close precipitate density and precipitate size, the sample studied in [14] had a lower in-field critical current density and a high critical current anisotropy. Thus Y₂O₃ precipitates alone cannot account for the strong pinning in our films.

Clues to the important defects may come from general observations of the dependence of J_{c-w} on our processing conditions. For thick films, J_{c-w} is generally higher when films are processed at higher YBCO growth rates and/or at lower temperatures [6]. For instance, J_{c-w} higher than 30 A mm⁻¹ in self-field could not be obtained by processing

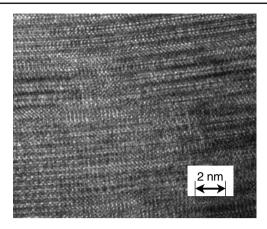


Figure 4. HRTEM cross-section of 3 μ m YBCO film on a buffered Ni–W substrate. The micrograph shows an area with considerable (00*l*) plane bending, which may be responsible for isotropic pinning shown in figure 2.

a 3 μ m-thick film above 735 °C or with a growth rate below 0.5 nm s⁻¹. Thus, fast growth rates and low reaction temperatures are two critical ingredients for achieving high J_{c-w} in these films. Since this processing environment is far from chemical equilibrium for the YBCO formation reaction but still sufficient to retain c-axis oriented grains, it is possible that small-scale lattice distortions are the pinning centres. This assumption was to some extent confirmed by high-resolution transmission electron microscopy (HRTEM) of a cross-section of a 3 μ m film processed under these conditions. The crosssection showed a relatively low density of stacking faults and multiple areas with localized bending of (00l) planes, a typical one being shown in figure 4. We speculate that by processing under non-equilibrium conditions (fast growth at low temperature) we quench this deformation, which otherwise would be relieved through formation of stacking faults. Such a deformation may be responsible for strong isotropic pinning and the practical absence of a $H \parallel ab J_c$ peak that we see in figure 2. Work is under way now to establish the statistical correlation between the amounts of deformation and the pinning.

An important advantage of these pinning centres is that there is little or nor degradation of $T_{\rm c}$. In our case $T_{\rm c}$ was 92.5 K, which is close to the highest ever reported for the YBCO compound. For example, additions of BZO to YBCO can dramatically improve the flux pinning, but there is a penalty in substantial reductions of $T_{\rm c}$, which can be as low as 86.2 K for an optimally doped sample [3]. Since critical current densities depend strongly on the temperature difference relative to $T_{\rm c}$, the tapes having large temperature margins are essential for reliable performance of electric devices operating at or near 77 K.

In summary we demonstrated a record high $J_{c-w}(H)$ by combining strong, isotropic flux pinning with high T_c in a 3 μ m-thick YBCO film manufactured by a scalable process. High growth rates of YBCO and low processing temperatures are two processing conditions essential for high J_c of the YBCO layer. There are two important implications of this work for the YBCO coated conductor technology: (1) it is possible to achieve strong isotropic pinning in

undoped thick YBCO layers, thus making additions of artificial defects unnecessary, and (2) isotropic in-field behaviour of these YBCO layers significantly simplifies the design of superconducting machines.

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References

- [1] Service R F 2005 Science 308 348
- [2] Coated Conductor Development Roadmapping Workshop II: Workshop Proceedings DOE/EE-000820404-CD accessible at http://www.eere.energy.gov/superconductivity/pdfs/ cc_workshopproceedings103_03final.pdf (accessed July 2004)

- [3] Kang S et al 2006 Science 311 1911
- [4] Foltyn R, Wang H, Civale L, Jia Q X, Arendt P N, Maiorov B, Li Y, Maley M P and MacManus-Driscoll J L 2005 Appl. Phys. Lett. 87 162505
- [5] Maiorov B, Wang H, Foltyn S R, Li Y, DePaula R, Stan L, Arendt P N and Civale L 2006 Supercond. Sci. Technol. 19 891
- [6] Solovyov V F, Wiesmann H J, Li Q, Welch D O and Suenaga M 2006 J. Appl. Phys. 99 13902
- [7] Feenstra R, Christen D K, Budai J D, Pennycook S J, Norton D P, Lowndes H H, Klanbunde C E and Galloway N D 1991 Proc. Sym. A-1, High Temperature Supercoducting Films at Int. Conf. on Adv. Mater. ed L Correra (Amsterdam: North-Holland) p 331
- [8] Solovyov V F, Wiesmann H J and Suenaga M 2001 Physica C 353 14
- [9] Goyal A et al 1996 Appl. Phys. Lett. 69 1795
- [10] Solovyov V F, Wiesmann H J, Wu L, Zhu Y and Suenaga M 2001 IEEE Trans. Appl. Supercond. 11 2939
- [11] Civale L, Marwick A D, Worthington T K, Kirk M A, Thompson J R, Krusin-Elbaum L, Sun Y, Clem J R and Holtzberg F 1991 Phys. Rev. Lett. 67 648
- [12] Yamada Y et al 2005 Appl. Phys. Lett. 87 132502
- [13] Palau A et al 2004 Appl. Phys. Lett. **84** 230
- [14] Ijaduola A O, Thompson J R, Feenstra R, Christen D K, Gapud A A and Song X 2006 Phys. Rev. B 73 134502